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IODINE INHALATION STUDY FOR PROJECT SEDAN

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Southwestern Radiological Health Laboratory
Las Vegas, Nevada

20 May 1964

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FOR

PROJECT SEDAN

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SOUTHWESTERN RADIOLOGICAL HEALTH LABORATORY
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IODINE INHALATION STUDY
FOR
PROJECT SEDAN
July 6, 1962

by
Southwestern Radiological Health Laboratory
U. S. Public Health Service

for
Nevada Operations Office
Atomic Energy Commission

Project Director
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Department of Health, Education, and Welfare
Public Health Service

AUTHOR'S NOTE

The subject matter of this report was presented under the title "Iodine Uptake by Dogs Exposed to Nuclear Cratering Cloud" at the Seventh Annual Western Industrial Health Conference in San Francisco, California on September 27, 1963 and under the title "Iodine Uptake From a Single Inhaled Exposure" at the Symposium on the Biology of Radioiodine at the Hanford Laboratories in Richland, Washington on June 17, 1963.

ABSTRACT

Beagle dogs and currently accepted physical air sampling equipment were exposed to the cloud produced by a nuclear cratering experiment to determine the deposition of radioactive iodine in organs of the biological sampler with that collected by the physical sampling devices. Primary emphasis is directed to the evaluation of such factors as isotopic ratios, rate of build-up, and the effect of the thyroid gland in concentrating iodine. The results, which indicated the selectivity of the biological sampler and the inefficiency of the physical samplers, are discussed.

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The U.S. Army, Veterinary Corps, supplied both personnel and equipment without which this project would have been impossible. Valuable personnel, equipment, and facilities support was also received from the University of Rochester, from Reynolds Electrical and Engineering Company, from the Southeastern Radiological Health Laboratory, and from other units of the Public Health Service. The U.S. Marine Corps supplied the helicopters and crews, without which the rapid retrieval of samples would have been impossible.

The support provided by these organizations and the helpful cooperation of the personnel assigned to this Study are gratefully acknowledged.

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Chapter 1

INTRODUCTION

Many studies both past and current have yielded reasonably reliable estimates of the parameters concerning transfer of iodine-131 through the food chain to the human thyroid. However, relatively little is known about the means by which radioiodine produced by nuclear testing passes through the biosphere to the food of cow and man, about its importance as an inhalation hazard, or about the relationship between its measurement in environmental samples or dose rate surveys to the radiation dose produced in man's thyroid. Project Sedan of the Plowshare Program provided a fission product source enabling some of these factors to be studied.

The U. S. Public Health Service, at the request of the Nevada Test Organization, Atomic Energy Commission, and with their financial support, designed and conducted a program to measure the inhalable fraction of iodine released to the close off-site area by the Sedan event. The nuclear device employed for the event was developed by the Lawrence Radiation Laboratory, Livermore, California. The device had a design yield of one hundred kilotons plus or minus ten per cent, and was buried at a depth of 635 feet in alluvium in Yucca Flat, Nevada.

It was expected that this iodine inhalation study would provide at least qualitative and hopefully some quantitative information concerning the effectiveness of a biological-physical field study in answering some of the urgent questions concerning health hazards of iodine, and would perhaps aid in a preliminary empirical evaluation of the relationship between external and internal beta-gamma doses which could result from tests producing a gaseous or near-gaseous nuclear cloud. It was understood that the information obtained would be unique, applying only to the conditions of the Sedan event. This unique character of the data became more evident when samples were received, as the expected energy spectrum of fission products was contaminated with large amounts of a gamma emitter (W^{187}) associated with the construction of the device. This contaminant masked many of the isotopes of interest in gamma spectrometric analysis of the samples.

A very short lead time allowed only limited calibration and field testing of many of the methods, equipment, and facilities being used in an experimental application for the first time. It was realized that this limitation, combined with that provided by the nature of the device, might render some of the data scientifically invalid. Nevertheless, since valuable experience and practical information were derived from the sampling and analytical procedures attempted, they are described whether or not they yielded scientific data in usable form.

Chapter 2

OPERATIONAL PLAN AND FIELD METHODOLOGY

One of the most persistent difficulties encountered in field experiments is that of effective sampler placement. The concept of manned mobile sampling stations was therefore employed to reduce the probability of missing the radioactive cloud without increasing the number of samplers required to obtain the desired information.

Samplers were concentrated in three mobile units carried in vehicles equipped with two-way radio. Each unit included two 2-wheel drive, four-speed transmission, air-conditioned panel trucks for transporting beagle dogs, and one 4-wheel drive truck carrying generators and physical sampling equipment. Each unit was manned by a team consisting of six members, two of which were responsible for biological sampling, two for physical sampling, one for continuous monitoring, and one to act as team historian. A supply team carrying fuel, water, and back-up equipment and supplies supported the three sampling units.

Placement of the sampling teams was directed by radio from the Nevada Test Site (NTS) Control Point. Standby positions were selected prior to the test day (D) to allow coverage of either of two parallel valleys

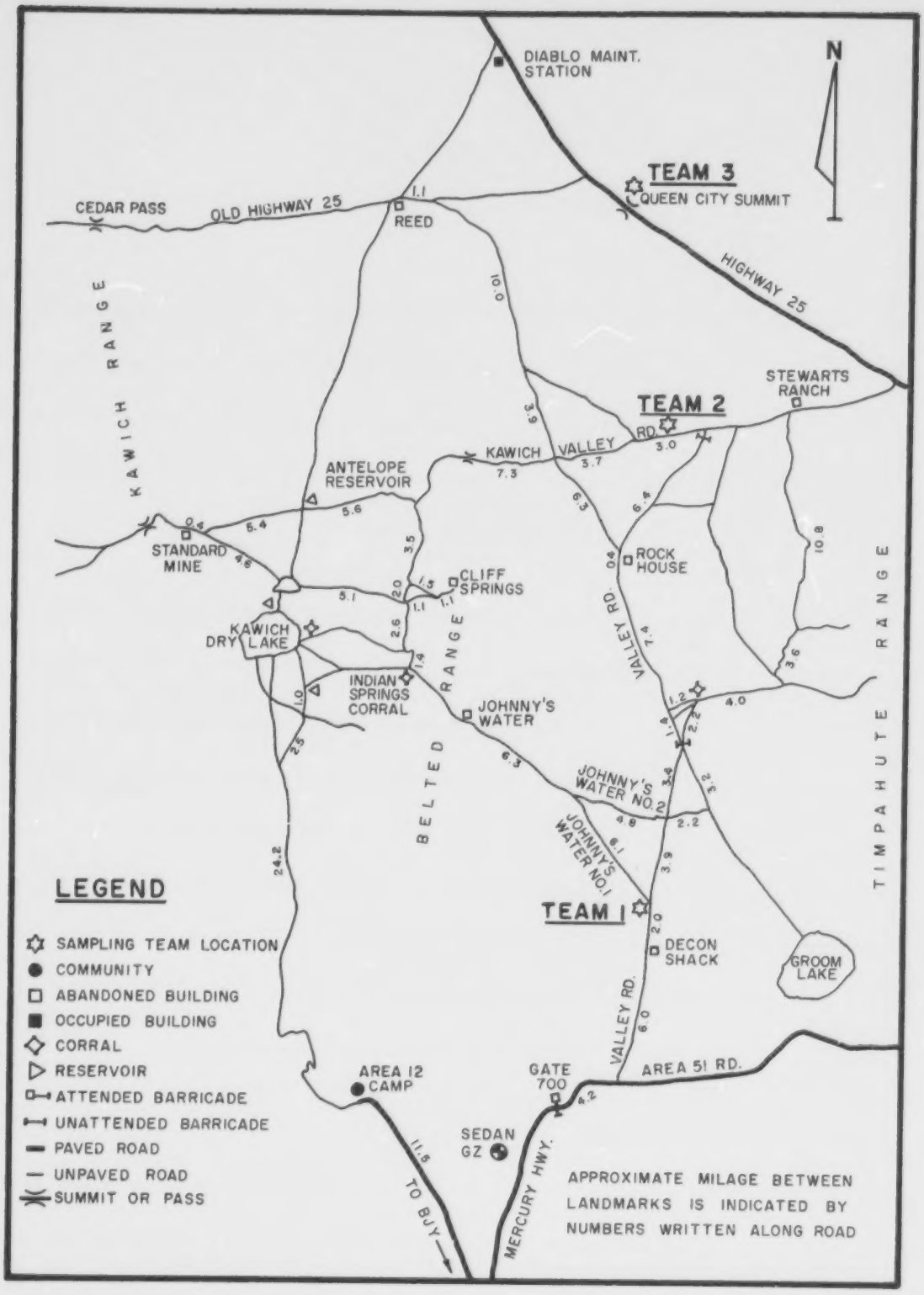

Figure 1. Map of the downwind area showing location of sampling teams.

within the predicted cloud trajectory. Road surveys and checks for radio contact were made prior to D-day in the area shown in Figure 1, and alternate sampling station approaches and escape routes were established. A complete dry run of the field operation on D-3 showed that only minor modifications of the plan were necessary.

Approximately 4 hours before detonation (H-4) the sampling and supply teams were dispatched to the pre-arranged standby positions. Since 35 minutes were required to place and set up a sampling station, just before detonation the team closest to ground zero (GZ) was directed by radio to set up at a position selected on the basis of U. S. Weather Bureau information. The remaining teams were positioned on the basis of information received from aerial cloud tracking and ground monitoring teams, as well as on the visual observations reported by the first sampling unit. The final sampling positions at approximately 14, 31, and 42 miles from ground zero are shown in Figure 1.

Sampling equipment was activated upon cloud arrival and was turned off when the cloud had passed, or as soon thereafter as possible. This minimized dilution with uncontaminated air, desorption of collected material, and contamination with resuspended material. Samples were packaged in the field and were picked up by helicopters summoned by each team as it began its shut-down operation. This allowed earlier and perhaps more significant analyses of short half-lived isotopes than would have been possible by the usual recovery methods. Two members

of each team accompanied the samples in the helicopter, while the four remaining members returned the sampling gear to headquarters in the three vehicles.

A facility for processing and analyzing samples was set up at the field headquarters to isolate the relatively "hot" samples obtained from this project from routinely analyzed low-level samples, and to allow as short a time lapse as possible between collection and analysis. The facility consisted of a compound enclosing two 40' x 8' volume van trailers housing counting room and radiochemistry laboratory, and three 50' x 10' office trailers serving as shops, treatment rooms for animals and office and storage space. This compound was conveniently located adjacent to a radiation safety facility maintained for the test site by Reynolds Electrical and Engineering Co. Helicopters and ground vehicles returning from the field were unloaded and surveyed in this rad-safe area. Samples and personnel were processed through standard rad-safe decontamination before entering the headquarters compound.

2.1 PHYSICAL SAMPLING

The physical sampling equipment at each station comprised four basic systems. These were the high and low volume air samplers, a cascade impactor, and a sequential sampler as described below.

High Volume Air Sampler. - 2 per station

This was a Staplex sampler equipped with 8" x 10" glass fiber prefilter and 3-1/4" diameter charcoal cartridge (MSA[1] Catalog No. CR-46727), drawing 25-35 cubic feet of air per minute as calibrated with a Venturi meter.

Low Volume Air Sampler. - 2 per station

This was a Gelman vacuum pump drawing air at about 0.7 cubic feet per minute through a 47 mm Millipore Type HA prefilter, a carbon cartridge consisting of a polyethylene tube 8" long by 9/16" I. D. backed by 8-14 mesh coconut shell activated carbon, and a flowmeter. A small-mesh screen wire and a spun glass plug were inserted into each end of the cartridge to hold the carbon in place. A Dwyer rotameter calibrated against a wet test meter was used to measure air flow. All connections were made with 1/4" Tygon tubing.

Cascade Impactor. - 1 per station

This was the standard Casella impactor for determining particle size, operated by a Gelman vacuum pump at a flow-rate of approximately 17 liters per minute. The four glass cover slip stages were backed by a Millipore filter fifth stage.

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[1] Mine Safety Appliances Co., Pittsburgh, Pennsylvania

Sequential Sampler. - 1 per station

This was the Gelman sequential sampler (Model No. 23000) with Whatman Type 41 filter paper, sampling in 10-minute periods for each cycle at a flow-rate of 16 liters per minute.

Each set of sampling equipment was mounted on a special wooden rack which allowed the entire physical sampling array for each station to be transported, set up, and operated as a single unit powered by three 1.5 kw generators. The rack was designed so that all samples were taken three feet above the ground.

When a station was shut down after passage of the radioactive cloud, all samples were packaged for helicopter transport to the field headquarters. The high volume prefilters were placed in glassine envelopes which were then taped shut. The high volume carbon cartridges were sealed in plastic bags. The cascade impactor was disconnected and sealed in a plastic bag. A plastic cover was placed over the low volume filter holder, and the filter holder and carbon cartridge were disconnected and sealed in separate plastic bags. The elapsed roll of filter paper from the sequential sampler was detached and sealed in plastic. All these bagged and sealed samples were then placed in a single large plastic bag for transport to the field headquarters.

2.2 BIOLOGICAL SAMPLING

Purebred beagle dogs were used as biological air samplers. These animals were obtained several weeks prior to the operation and were housed in kennels at the headquarters compound. To quantitate air sampled by the dogs, and to relate the radioiodine collected with that collected by the physical air sampling systems, respiratory frequency and inspired air volume of each animal were determined before the Sedan event. The measurements were made under simulated field conditions after the animals were acclimated to summer desert conditions. The measurement system utilized Fleisch Pneumotachographs[1] of various flow capacities, built into latex-coated plaster of paris face masks individually constructed and fitted to each dog. Pressure differences sensed by the pneumotachograph were converted through a battery operated Sensitive Differential Pressure Transducer, Model 1004A[2] containing the transducer, a control indicating meter, and a recorder driver, to a tracing on the high torque, spring operated Model AW Esterline Angus recorder.

To supplement these pre-event breathing calibrations, two animals at each sampling station wore the breathing apparatus during cloud passage. After the event, respiratory measurements were continued on all dogs not sacrificed immediately after exposure.

Since time was not available to train the dogs in sampling behavior,

[1] Instrumentation Associates, New York
[2] Monroe Electronic Laboratories, Inc., Middleport, New York

they were exposed in cages made of wire mesh. All animals were prepared and caged at the headquarters compound. Two air-conditioned panel trucks carried ten caged dogs to form the biological sampler complement at each sampling station.

It was intended that each dog be tranquilized by oral administration of chlorpromazine upon arrival at the sampling locations to facilitate handling. However, difficulties encountered in administering the tranquilizer prevented tranquilization of animals at the station closest to GZ and resulted in partial tranquilization of animals at the other two stations. Breathing measurement apparatus was connected to two dogs at each station to operate throughout the sampling period as reference measurements. All dogs were placed on a platform three feet above the ground in close proximity to the physical sampling devices.

To determine the body burden of activity inhaled during cloud passage, it was essential to prevent inhalation of resuspended material and ingestion of material deposited on the nose and fur. Therefore, four animals at each station, including one of those wearing the respiratory measurement devices, were sacrificed as soon as possible after cloud passage. The area surrounding the nose and mouth of each remaining animal was washed with a detergent solution to prevent introduction of additional activity by lapping. The dogs were left in their restraining cages to prevent lapping of other areas of the body. After intravenous administration of sodium pentobarbital, sacrifice was accomplished by maximal blood withdrawal from the heart, using standard blood donor

kits to withdraw and receive the blood. Sacrificed animals were sealed in large polyethylene bags before being loaded into the helicopters with the caged dogs, the blood samples, and the physical samples for transport to the headquarters compound. Before being readmitted into the compound, all dogs were decontaminated at the Area 400 Rad-Safe facility with warm water and detergent, and the neck and chest areas were shaved. The sacrificed animals were again sealed in clean polyethylene bags before being taken to the clean area, and the living animals were returned to the kennels to await in vivo counting of the thyroid and serial sacrifice.

2.3 DOSIMETRY

Dose rate readings were taken at each station with portable survey instruments including the Beckman MX-5 (range 0-20 mr/hr), the Eberline E500-B (range 0-2 r/hr), and the Tracerlab T1B (range 0-50 r/hr). Readings were recorded every ten minutes until the cloud arrived and every three minutes during cloud passage. Integral gamma dose to each station was determined from Du Pont Type 556 film badge dosimeters containing high and low range film components. These badges were attached to the physical sampling gear racks.

One roentgen and 10 roentgen ionization chamber dosimeters and high and low range film badges were used as personnel dosimeters. These were standard personnel dosimeters issued and analyzed by the REECo Radiation Safety Dosimetry Section. The protective clothing used was

also supplied by REECo Rad-Safe and consisted of standard radex suiting in addition to respirators with charcoal cannisters. Team members suited up in the field just prior to cloud arrival. Respirators were worn only when the dose rate rose to a pre-determined level. Upon return from the field, team personnel were decontaminated by standard procedures at the REECo Rad-Safe facility.

Chapter 3

LABORATORY METHODOLOGY

3.1 PROCESSING OF SAMPLES

For the most part, processing of samples prior to counting consisted of simply repackaging the samples in clean containers when they had returned from the field. Chemical separation of iodine was performed only on low volume sampler prefilters twelve days after the event. Particle sizing attempts were not successful.

Dog autopsies were performed through the polyethylene bags to minimize contamination of internal tissues. A midline incision was made through the bag and skin from larynx to sternum. A skin-and-bag flap was reflected to expose neck musculature, which was then dissected with a clean set of instruments to expose the thyroid gland, trachea, and esophagus. With a third set of clean instruments the thyroid gland was carefully removed to a small plastic bag in which it was weighed and counted. The trachea was reflected and the esophagus removed and sealed in a plastic bag, again using clean sets of instruments for each operation. After continuing the midline incision from sternum to pubis, further dissection was carried out in an anterior to posterior

direction to remove respiratory system, stomach, small intestine, large intestine, kidneys, and gonads. Each of these samples was sealed in a pre-weighed plastic container in which its weight and redioactivity was determined. Extreme care was taken throughout the sample processing procedures to prevent redistribution of activity or cross contamination of samples.

3.2 RADIOASSAY OF SAMPLES

Three systems were used for assaying the gamma activity of samples. The systems were assembled from components on hand, borrowed from routine programs, or purchased new if time permitted. The urgency of the Sedan program, the short lead time, and the necessity for returning many system components to their routine duties at other locations limited the extent of calibration. Also, it was not possible to acquire the desired spectrum of gamma energies from the standards available. The three gamma analysis systems are described below and the standards used for their calibration are listed in Table 1.

Table 1. List of radioisotope standards.

ISOTOPE	HALF LIFE	ENERGY
I^{131}	8.08 days	0.36 Mev 0.64 Mev
Cs^{137}	26.6 years	0.66 Mev
Zn^{65}	245 days	1.12 Mev
K^{40}	1.25×10^{9} years	1.46 Mev

Calibration curves for two of the systems are shown in the Appendix, Part B.

System 1. 4" x 4" NaI(Tl) crystal assembly with RIDL 400-channel gamma pulse height analyzer.

Most samples analyzed by this system were sealed in the quart size plastic cheese-tub type container having a 3-3/4" diameter base. To calibrate for this configuration, counting efficiencies were determined for various levels of a solution of each standard. Channel width was set to be 10 Kev. For photopeak energies below 900 Kev, counts were summed over eight channels (80 Kev). Above 900 Kev, nine channels (90 Kev) were summed. The counting efficiencies obtained are shown in Table 2.

Samples in three additional configurations were analyzed with the 4" x 4" scintillator-spectrometer system. These were the MSA charcoal cannister, the hand-packed tube of charcoal, and the membrane filter from Gelman low volume samplers.

To calibrate for the MSA cannister, a portion of charcoal equivalent to approximately 1/8" penetration was removed and impregnated with a known volume of the iodine-131 standard solution. After slow drying, the impregnated charcoal was mixed with the uncontaminated granules and replaced in the cannister.. Both the cannister and the container used for drying the charcoal were counted, showing a 16% detection efficiency for I^{131} in the cannister configuration.

Table 2. Efficiencies of the 4"x 4" solid crystal of System 1 and the 9"x 8" well crystal of System 2 for counting radioisotope standards.

ISOTOPE STANDARDS	SYSTEM 1.				SYSTEM 2.			
	STD. GEOMETRY		RANGE SUMMED (Kev)	COUNTING EFFICIENCY (%)	STD. GEOMETRY		RANGE SUMMED (Kev)	COUNTING EFFICIENCY (%)
	VOL. (ml)	HT. (in)			VOL. (ml)	HT. (in)		
I^{131}	115	3/4	80	13.5	200	2	80	55.9
	230	1-3/8	80	10.5			90	59.0
	345	2	80	8.62				
	575	3-1/2	80	6.24				
Cs^{137}	115	3/4	80	14.1	200	2	80	57.5
	230	1-3/8	80	11.4			90	62.9
	345	2	80	9.25				
	460	2-3/4	80	7.80				
Zn^{65}	115	3/4	90	5.25	200	2	110	27.5
	230	1-3/8	90	4.20			130	30.9
	345	2	90	3.40				
	460	2-3/4	90	2.92				
K^{40}	115	3/4	90	2.05	200	2	110	18.3
	230	1-3/8	90	1.68				
	345	2	90	1.32				

In a similar manner the charcoal from the hand-packed tubes was calibrated for counting in the quart-sized tub. A detection efficiency for I^{131} in this configuration was also found to be 16%, as it was for the membrane filter placed in a 2" diameter stainless steel planchet, impregnated with the standard I^{131}, and counted on top of the crystal.

System 2. 9" x 8" NaI(Tl) crystal assembly containing 3" x5" well with RIDL 400- channel pulse height analyzer.

A snap-top plastic container having a flat base and a very slight taper, custom-made by Nalge Co.[1] to fit the crystal well, was not available for use during the Sedan event. Therefore, all samples analyzed in this system followed the geometrical configuration of a 500 ml. plastic bottle 2-3/4" diameter and 6-1/2" in height. A 200 ml. volume rising approximately 2" above the container base was selected as a volume representative of the samples to be counted in this system. The resolution of this crystal was poorer than that of the 4" x 4", spreading photopeaks over a wider range. Therefore, efficiencies were determined by summing from eight to thirteen channels depending on photopeak energy. This is shown in Table 2 with the detection efficiencies obtained.

[1] The Nalge Company, Rochester, New York

System 3. 5" x 6" NaI(Tl) crystal with 3" x 5" well and RIDL scaler.

This system was used to determine gross gamma activity. Sample geometry was similar to that in the 9" x 8" well crystal assembly. Two hundred milliliters of standard solution in 500 ml. polyethylene containers were counted and the efficiencies calculated. In addition, an efficiency was calculated for 200 ml. of a mixed standard solution. Since efficiency is highly dependent on gamma energy, it was realized that the method would not give a true efficiency for mixed fission products. True efficiency would depend on the relative levels of activity at the various energies. Since it was expected that the lower energy isotopes would be more prevalent, gross gamma detection efficiency was determined for a mixture of I^{131} and Zn^{65} standard solutions in which the activity of I^{131} was greater by a factor of three. This efficiency was found to be 63%.

Physical samples received for counting were MSA charcoal cannisters, hand-packed charcoal tubes, membrane prefilters, 8" x 10" glass fiber prefilters, and the Millipore filters and glass cover-slip stages from cascade impactors. Gamma pulse height analysis of these samples could not be made immediately because of the high levels of radioactivity they contained and the complexity of the isotope mixture. Low volume sampler membrane and charcoal filters were analyzed prior to

D+5, but at D+5 it was still impossible to obtain information on the high volume samples. By D+28 the activity in the high volume samples had decayed to countable levels.

An attempt was made to count gross beta activity on the membrane prefilters and impactor cover-slips by placing each sample on a stainless steel planchet and counting it in an internal proportional counter. However, dust particles were blown around the chamber during the gas purge, causing gross contamination of the equipment. Therefore, these samples were sealed in polyethylene containers and counted for gross gamma activity only.

The biological samples received for counting included thyroid, blood, respiratory system, esophagus, small intestine and contents, large intestine and contents, kidneys, and gonads from each dog. Gross gamma activity in these samples was determined as soon as possible after autopsy. These data were to be used if a gamma pulse height analysis of each organ could not be made, since it was possible to estimate from the gamma scans the percentage of gross gamma activity contributed by the various isotopes of the mixture. By this method, a quantitation of the isotopes present was obtained in some organs which had not been gamma scanned. The gross gamma count was also used to indicate the level of activity on the sample before a detailed pulse height analysis was attempted.

One- to five-minute gamma scans were made of all organs except those so highly contaminated as to produce approximately 100% analyzer dead time. The larger organs were placed in the tub container and scanned on the 4" x 4" crystal. The smaller organs were scanned in the well crystal. A second scan was obtained on all organs one or two days later and, when possible, a third count was made several days later. All the thyroid samples were recounted on D+5 and again on D+22.

3.3 IN VIVO ANALYSIS

In addition to serial sacrifice and autopsy of exposed dogs followed by in vitro analysis of organs and tissues, an attempt was made to follow the build-up and decay of iodine in the thyroid by in vivo counting. To do this, a system was constructed in which two 3" x 3" NaI(Tl) crystals were optically coupled to two photomultiplier tubes, which in turn were coupled to preamplifiers feeding into the two inputs of a TMC(400-channel) pulse height analyzer. Each crystal was encased in a 1-1/2" lead flat-field collimator. In addition, a 2" x 2" NaI(Tl) crystal encased in 1/2" lead sheet was available as a supplementary detector.

Meaningful calibration, effective collimation, and reproducible geometry are necessary features of an in vivo counting system. A reproducible procedure for the Sedan study was established by making several trial counts with the available equipment, but insufficient time was available for further refinements. One of the dogs which had not been

exposed served as a control. The greatest reproducibility was obtained in trial counts by placing one 3" x 3" crystal to view the right side of the neck and the other to view the thyroid from underneath. By using this method to scan the thyroids of exposed dogs, barely detectable levels of radioiodine were seen, although tellurium-132 and tungsten-187 were present. This indicated that the flat-field collimator had allowed the crystals to view more than just the thyroid (probably the respiratory system). Therefore in vivo counting data were used only to indicate the relative change in iodine content of the animal thyroid as a function of time.

Table 3. Qualitative analysis of the body burden of gamma emitters in a dog sacrificed 24 hours after inhalation exposure 42 miles from ground zero.

SAMPLE ANALYZED	DAY ANALYZED	ISOTOPES DETECTED
Thyroid Gland	D+2	I^{131} I^{133} Xe^{133m}
Respiratory System	D+1	W^{187} Te^{132} I^{133} Xe^{133m}
Esophagus	D+2	W^{187} Te^{132} I^{131} I^{133} Xe^{133m} Xe^{135}
Small Intestine and Contents	D+1	W^{187} Te^{132} I^{133} I^{131} Xe^{133m}
Large Intestine and Contents		Activity exceeded counting capacity
Kidneys	D+1	W^{187} Te^{132}
Gonads (testes)	D+1	W^{187}
Blood	D+1	W^{187} Te^{132} I^{131} I^{133}

Chapter 4

RESULTS

The high levels of activity, the complexity of gamma spectra, the presence of unusual contaminants such as W^{187}, and the minimal calibration of counting equipment all contributed to the difficulty of making a valid analysis of the data, and reduced the reliability of much of the data obtained. A discussion of the data analysis procedure and a sample calculation are included in the Appendix. The results presented in this section are considered to be reliable for relative comparisons, for order of magnitude quantitation, and for indicating trends. In view of the many imponderables, no attempt has been made to determine probable error.

A qualitative analysis of the organs of a dog which had been exposed at the 42-mile station and sacrificed twenty-four hours later showed the body burden contained, at time of count, the isotopes listed in Table 3. Because of the complexity of the spectra of the various organs, it was decided to consider the thyroid data primarily. Since only radioiodine and its daughters are expected in the gamma spectrum of an exposed thyroid, quantitative estimates could be made. Initial count, recount after five days, and a third count after twenty-two days would assure a

Table 4. Data from biological samplers, including iodine in dog thyroids analyzed in vitro.

EXPOSURE LOCATION AND TIME OF CLOUD ARRIVAL	SACRIFICE TIME (±1/2 hr)	DOG NO.	AVERAGE RATE OF INHALATION (ml/min)	VOLUME OF AIR INHALED (M^3)	NET dpm IN THYROID extrapolated to sacrifice time	
					I^{131}	I^{133}
Station 1		11	818	6.13×10^{-2}	- - - - -	- - - - -
		16	2375	17.8×10^{-2}	- - - - -	- - - - -
GZ +14 miles	H + 1-3/4	17	744	5.58×10^{-2}	- - - - -	- - - - -
		43	2153	16.1×10^{-2}	- - - - -	- - - - -
		22	908	6.81×10^{-2}	- - - - -	- - - - -
H + 1/2	H + 29	27	1012	7.59×10^{-2}	- - - - -	- - - - -
		37	946	7.09×10^{-2}	8.00×10^{2}	- - - - -
		2	1167	8.75×10^{-2}	4.59×10^{3}	- - - - -
	H + 73	3	794	5.95×10^{-2}	1.47×10^{3}	- - - - -
		28	1590	11.9×10^{-2}	2.38×10^{3}	- - - - -
Station 2		4	703	9.84×10^{-2}	- - - - -	3.05×10^{4}
		5	368	5.15×10^{-2}	4.24×10^{3}	1.35×10^{5}
GZ +31 miles	H + 1-1/4	14	741	10.4×10^{-2}	6.74×10^{3}	1.88×10^{5}
		23	750	10.5×10^{-2}	2.83×10^{3}	5.90×10^{4}
		15	1462	20.5×10^{-2}	6.43×10^{5}	6.14×10^{6}
H + 1-1/3	H + 30	33	1558	21.8×10^{-2}	6.75×10^{5}	6.21×10^{6}
		38	980	13.7×10^{-2}	2.97×10^{5}	2.74×10^{6}
		21	1058	14.8×10^{-2}	6.52×10^{5}	1.36×10^{6}
	H + 75	26	1507	21.1×10^{-2}	8.67×10^{5}	1.59×10^{6}
		32	1262	17.7×10^{-2}	6.00×10^{5}	1.73×10^{6}
Station 3		7	685	5.96×10^{-2}	- - - - -	- - - - -
		12	1056	9.19×10^{-2}	5.95×10^{3}	1.37×10^{5}
GZ +42 miles	H + 3-1/2	30	1324	11.5×10^{-2}	1.09×10^{3}	2.96×10^{4}
		41	2242	19.5×10^{-2}	- - - - -	- - - - -
		25	1501	13.1×10^{-2}	2.29×10^{5}	2.33×10^{5}
H + 2	H + 28	35	1248	10.9×10^{-2}	6.44×10^{4}	6.60×10^{5}
		40	1756	15.3×10^{-2}	7.50×10^{4}	7.57×10^{5}
		8	1119	9.73×10^{-2}	1.24×10^{5}	2.58×10^{5}
	H + 75	18	629	5.47×10^{-2}	1.26×10^{5}	2.75×10^{5}
		24	996	8.66×10^{-2}	3.13×10^{5}	6.85×10^{5}

good quantitation of some of the radioiodines present.

Table 4 presents the in vitro iodine analyses of dog thyroids. The volume of air sampled by each dog, as determined by the method described, is also listed. It can be seen that the ratio of I^{133} to I^{131} decreases with the length of time before sacrifice. The ratios are the same for a given sacrifice time at both stations, indicating that no appreciable fractionation of iodine or its precursors occurred as the cloud moved from thirty-one to forty-two miles.

Table 5 presents the results of the analyses of the low volume air samples. No I^{131} was detected in these samples, indicating the relatively negligible amount of that isotope present in the air at the times of sampling. When the activity of I^{133} per cubic meter of air determined from the low volume samples is compared with the I^{133} activity per cubic meter of air breathed by each dog sacrificed immediately after exposure, the values of I^{133} concentration in air are essentially the same.

Table 5. Iodine collected by low volume air samplers.

Sample Location	Collector Type	Flow Rate (cfm)	Air Sampled (M^3)	I^{133} Activity* (dpm/M^3)	Total I^{131} ** Activity* (dpm/M^3)
Station 1 GZ +14 mi.	Membrane	.71	1.47	7.27×10^3	2.09×10^4
	Charcoal			1.26×10^4	
Station 2 GZ +31 mi.	Membrane	.54	2.07	1.07×10^6	1.22×10^6
	Charcoal			1.55×10^5	
Station 3 GZ +42 mi.	Membrane	.61	1.25	1.31×10^6	1.40×10^6
	Charcoal			8.65×10^4	

*Activity has been extrapolated to mid-sampling time.
**Iodine ^{131} measured on both membrane and charcoal filters.

Plots of the initial in vivo counts indicated a barely detectable amount of radioiodine. This was due to the time lag between inhalation and deposition in the thyroid. However, Te^{132} and W^{187} were present in these initial plots, indicating that the flat-field collimator had viewed more than just the thyroid. Therefore, these data were used only to indicate the change in iodine content of the animal thyroid as a function of time. A smaller crystal with a higher degree of collimation would have yielded more useful information.

Figures 2 and 3 are typical spectra obtained from in vivo counting of a dog from the 31-mile and one from the 42-mile station at the time and dates indicated. The change in iodine content of the thyroids is apparent. It must be emphasized that the change pictured does not apply specifically to the thyroid, because other portions of the animals' bodies contributed to the spectra obtained. The qualitative picture shown was substantiated by the in vitro results.

The majority of the individuals assigned to field teams wore protective respiratory equipment during cloud passage, or were evacuated from their stations. One person, however, remained at the 42-mile station during the entire one-half hour period of operation. He wore no respirator at any time, although he was in and out of his vehicle. Shielding afforded by the vehicle reduced his external gamma exposure to 525 mr as compared to the total station exposure of 870 mr. On D+7, an energy spectrum was determined on this individual at the Walter Reed Army

Institute of Research in their Whole Body Counting Facility. A 9" x 4" sodium iodide crystal, and calibration procedures and efficiency values obtained from counts done on children were used. At the time of this analysis, the total thyroid burden of I^{131} was calculated to be between 0.1 and 0.3 microcuries. On D+1, a thyroid scan was obtained on this same individual using the equipment employed for in vivo counting of dogs. The I^{133} thyroid burden was found to be 0.16 microcuries corrected to mid-time of exposure.

A second individual at the same (42-mile) station wore protective respiratory equipment throughout the entire time of cloud passage. His external gamma exposure was 475 mr. On D+12, he was examined in the whole body counting facility at New York University. His thyroid burden of I^{131} on D+12 was 8×10^{-4} microcuries.

On D+12, one of the individuals assigned to the 31-mile station was also examined in the whole body counting facility at New York University. His body burden of I^{131} was 1×10^{-3} microcuries on D+12. However, he wore a respirator throughout the entire period except for that portion of the time between team evacuation and return for sample pick-up. His estimated time at the station was twenty-seven minutes prior to evacuation, during which time his external gamma exposure was 1025 mr compared to a station exposure of 2.9 roentgens.

A fourth individual, a member of the Off-Site Radiological Safety Program, was examined on the local in vivo system on D+1. His I^{133}

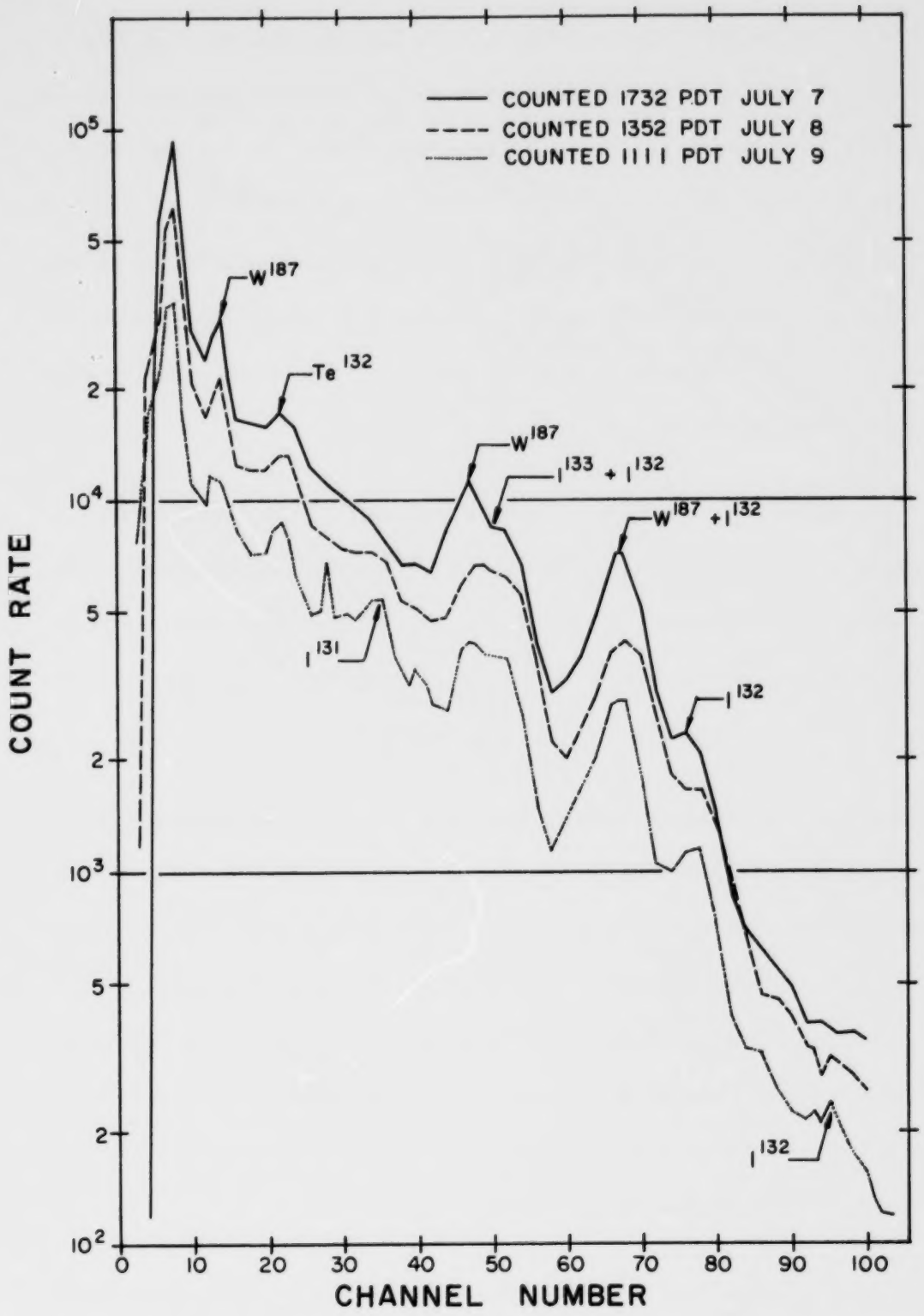

Figure 2. In vivo spectra of the thyroid of one dog from the 31-mile station.

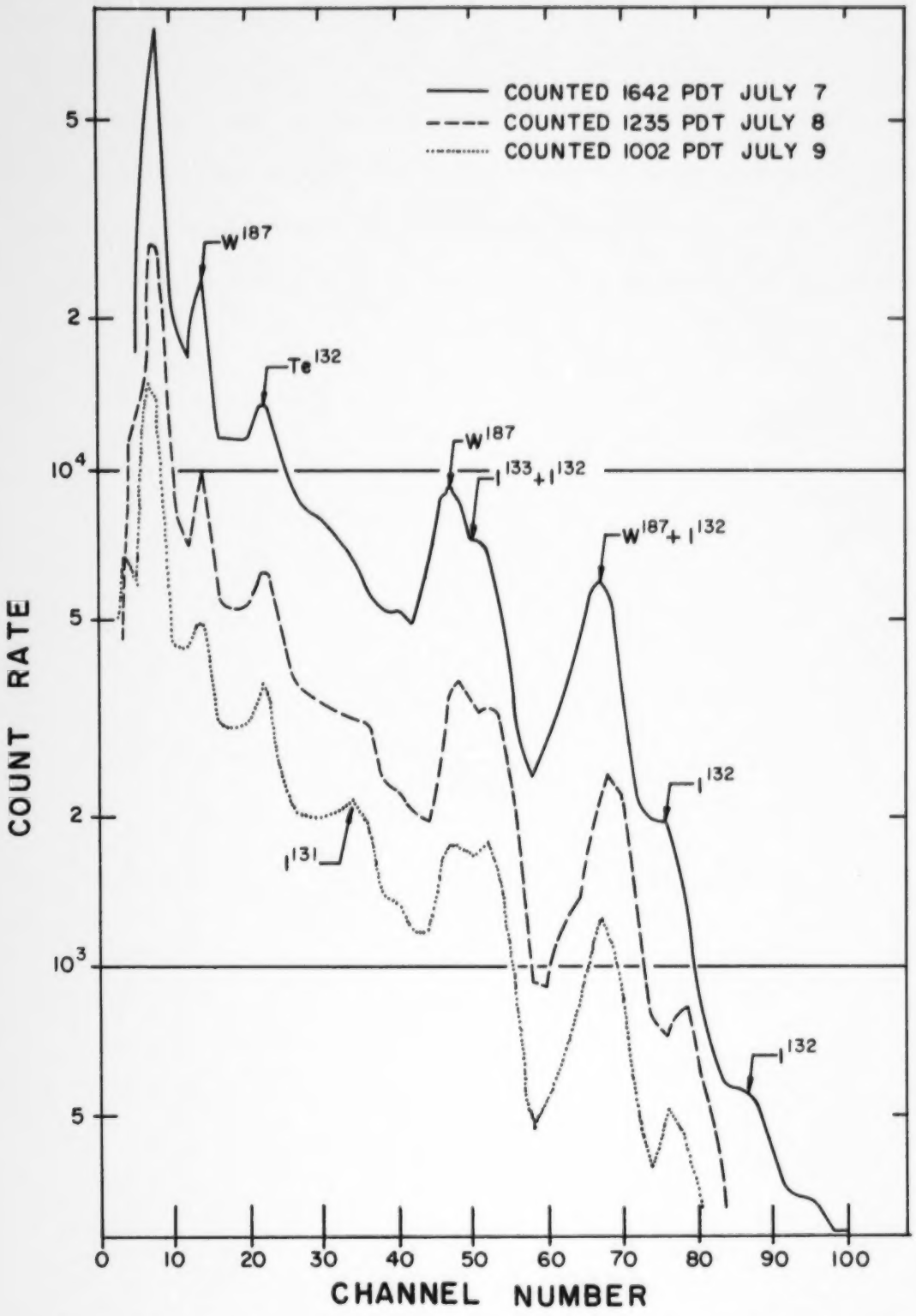

Figure 3. In vivo spectra of the thyroid of one dog from the 42-mile station.

thyroid burden was estimated to be 1.4×10^{-2} microcuries corrected to mid-time of exposure. This person's duties took him back and forth through the cloud in the vicinity of the 42-mile station and, although his duration of exposure is not known, it has been established that no protective breathing equipment was worn. His external gamma exposure was 770 mr.

Chapter 5

CONCLUSIONS

A comparison of the average amount of I^{133} collected by the low volume samplers (Millipore prefilters plus activated carbon cartridges) with that found in the thyroids of dogs sacrificed immediately after cloud passage is of interest. At the 31-mile station, the low volume sampler showed the average I^{133} content of the cloud to have been 1.2×10^6 dpm per cubic meter (see Table 5). Four dogs were sacrificed at that station, and their thyroids were removed and counted separately. The average value obtained from counting the four thyroids indicated an I^{133} content of 1.4×10^6 dpm in the thyroid per cubic meter of air inhaled. This concentration value is obtained by dividing total I^{133} in each thyroid by the total amount of air breathed (sampled) by each dog. A similar agreement existed at the 42-mile station. Here, the low volume air sampler indicated an I^{133} content in air of 1.4×10^6 dpm per cubic meter, and the average amount found from counting the thyroids of two dogs was 0.9×10^6 dpm per cubic meter.

Two interpretations of these data are possible. First, it could be assumed that all I^{133} inhaled by a beagle dog goes directly and rapidly to the thyroid and that the physiological system is a completely efficient

sampling mechanism. This conclusion is quite improbable. One must therefore accept the second interpretation and assume that the physical sampling gear currently considered optimum for sampling radioactive iodine effluents is actually very inefficient, being on the order of 10% or less. Thus, there is an urgent need for additional studies directed toward the determination of radioiodine concentration in air which will lead to the development of truly quantitative sampling methodology.

Of all the radioactive iodine isotopes, I^{131} has justifiably received the greatest attention. This is logical when one considers ingestion alone at times after a release measured in days. The situation is entirely different when relatively near distances and shorter times are considered for inhaled material. No I^{131} was detected in either the prefilters or the activated carbon cartridges at the 31-mile or 42-mile stations when they were counted immediately upon return to the laboratory. The filters were stored, however, and iodine was extracted chemically at D+12 and analyzed for I^{131}. The I^{131} was then extrapolated back to the mid-time of the sampling period to give a hypothetical I^{131} content of the cloud. At the 31-mile station, the hypothetical I^{131} concentration in air was 1.2×10^6 dpm per cubic meter, and at the 42-mile station was 1.4×10^6 dpm per cubic meter. Admittedly, these data are subject to large errors due to such factors as sampler inefficiencies and incomplete chemical extractions. It may be significant, however, that the averages of the animal thyroids from the 31- and 42-mile stations indicated respectively 5.9×10^4 and 3.7×10^4 dpm of I^{131} per cubic meter.

The presence of I^{131} is certainly confirmed in the cloud, and is in agreement with the theoretical fact that at the time of sampling, I^{131} represents 0.6% of the total iodine activity and I^{133} represents 11%. There was probably very little I^{131} on the physical samples counted immediately, and that obtained by chemical extraction at D+12 resulted from the decay of Te^{131} captured on the filters. It can be concluded that the animal takes a more accurate representation of the iodine activities than does physical sampling equipment because the animal "does its own chemistry" and deposits these activities in strictly correct ratios in the thyroid gland.

A comparison of the I^{133}/I^{131} ratio found in the dog thyroids with theoretical ratios as a function of time supports the validity of this conclusion. At H+1.5 the I^{133}/I^{131} ratio in the thyroids at the 31-mile station was 23.7. At the 42-mile station the ratio was 24.4. According to Glendennen's theoretical calculations, the ratios are 24.4 at one hour and 19.6 at three hours after instantaneous fission of U^{235}.

No obvious correlation or trend was observed between the ratio of total radioactive iodines in animals' thyroids compared with the total external gamma dose at either the 31- or 42-mile stations.

APPENDIX

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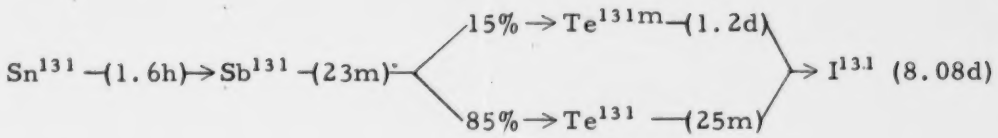
Part A

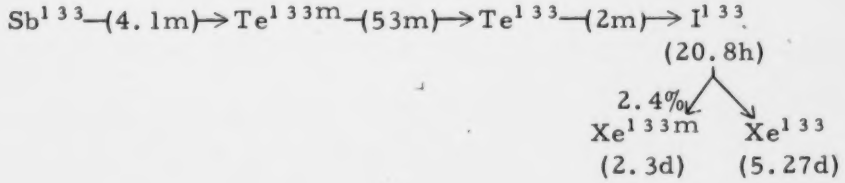
TABLE OF DECAY SCHEMES



Chain Member	Energy (Mev)
Te^{131m}	.18, .84, .77
Te^{131}	.15, .94, .45
I^{131}	.36, .64

Sn^{132} —(2.2m)→ Sb^{132} —(2.1m)→ Te^{132} —(77.7h)→ I^{132} (2.3h)

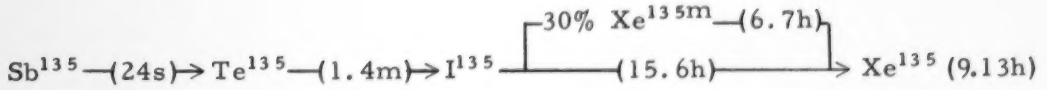
Chain Member	Energy (Mev)
Te^{132}	.23
I^{132}	.67, .78, .53, .96, 1.40



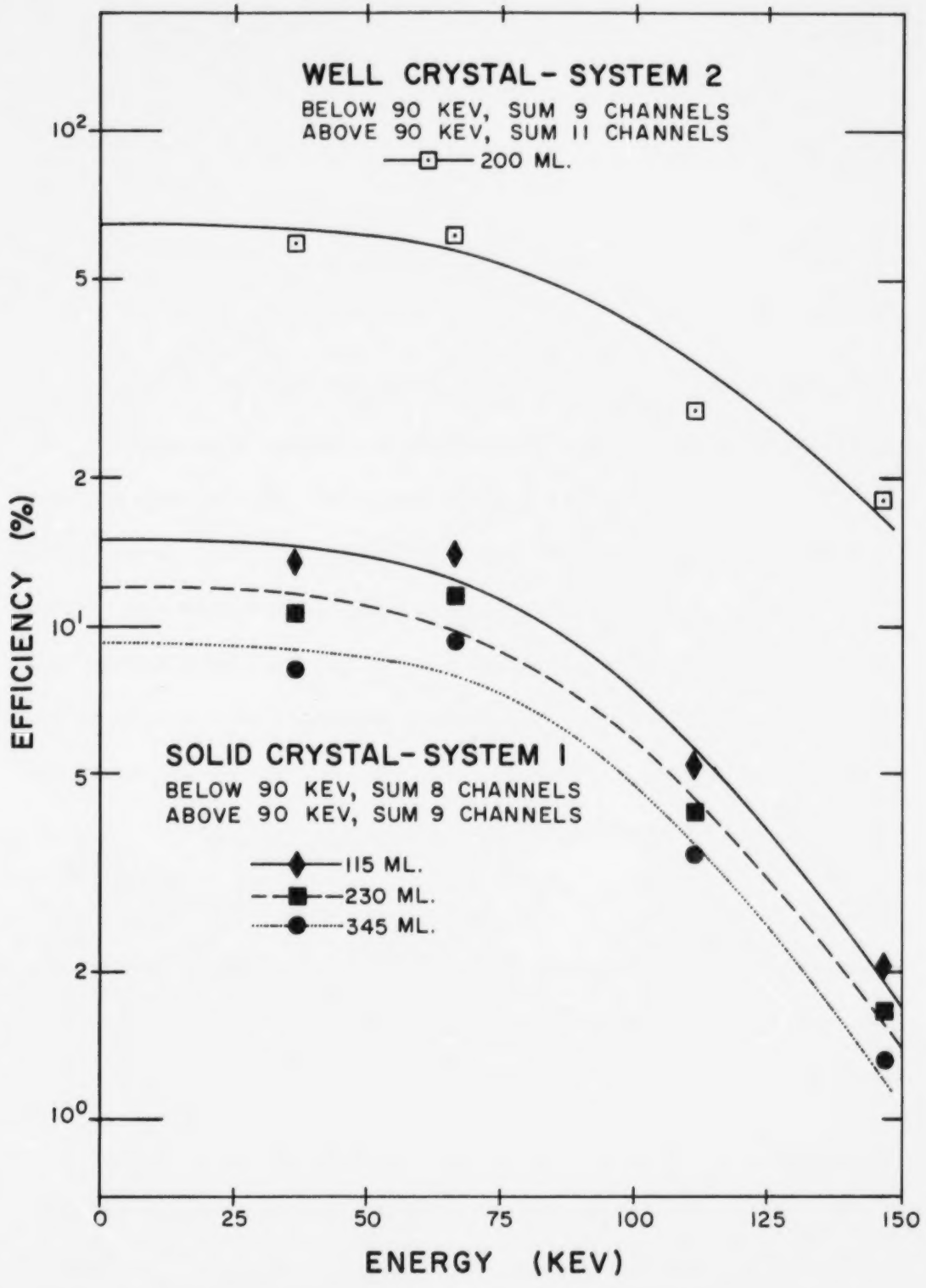
Chain Member	Energy (Mev)
Te^{133m}	.33, .40
Te^{133}	.60, 1.00, .40
I^{133}	.53, .85, 1.40
Xe^{133m}	.23
Xe^{133}	.08

$Sb^{134} \xrightarrow{(48s)} Te^{134} \xrightarrow{(42m)} I^{134}$ (53m)

Chain Member	Energy (Mev)
I^{134}	1.10, .86, .12, .20, 1.78



Chain Member	Energy (Mev)
I^{135}	1.14, 1.28, .53, 1.72, 1.46, .86, 1.80, 1.04, .42
Xe^{135m}	.23
Xe^{135}	.08



Part B. Calibration curves for two gamma pulse height analysis systems.

Part C

DATA ANALYSIS

C-1. DETERMINATION OF IODINE CONCENTRATION IN AIR

In determining the iodine content of the air during cloud passage, the data from the low volume samplers were used. The activity on these samples was low enough to allow several gamma scans to be made. Also, chemical separation of the iodine from the prefilter was carried out. Two extractions were made on the prefilters approximately five days apart. The iodine from the first extraction is a function of the quantity sampled and grown in to H+7, whereas the second extraction indicates ingrowth alone.

The gamma scan from D+4 was used in calculating the I^{133} in the charcoal filter. The contribution to this energy region from other photopeaks was again assumed to be negligible. Calculation of I^{131} was carried out by relating gamma scans from D+27 to data from D+4.

The quantity of iodine calculated from the membrane filter is the combined activity of the actual iodine sampled and that produced by decay of its precursors. To determine how much actual iodine was present in the air at the time of sampling, a different approach was taken.

Extrapolation of the measured value back to time of sampling is not valid, since the value actually represents decay and ingrowth of the isotopes.

Correction for this was made through use of the information provided by the two chemical extractions on the prefilter. By considering the formation of radioactive daughters in the decay of the parents for an n-member decay chain, a relationship can be established, for any time, between the existing quantity of an isotope, its initial quantity, and the quantity of its ancestor. This is a straightforward application of the Bateman equation[1].

The Bateman solution for a chain of n members in which only the parent substance is present at t_o is:

$$N_n = C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + \ldots\ldots\ldots C_n e^{-\lambda_n t}$$

Where,

$$C_1 = \frac{\lambda_1 \lambda_2 \ldots\ldots\ldots \lambda_{n-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1) \ldots\ldots (\lambda_n - \lambda_1)} N_1^o$$

$$C_2 = \frac{\lambda_1 \lambda_2 \ldots\ldots\ldots \lambda_{n-1}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2) \ldots\ldots (\lambda_n - \lambda_2)} N_1^o, \text{ etc.}$$

If more than just the parent substance is present at t_o, an addition is made to the above solution; a Bateman solution for an (n-1)-membered chain with substance 2 as the parent, a Bateman solution for an

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1 Friedlander, G., and J.W. Kennedy, Nuclear and Radiochemistry, John Wiley & Sons, New York, 1955, p. 36.

(n-2)-membered chain with the next substance as the parent, etc. A calculation of the percentage Te^{131} in air is given in Part D as an example of a Bateman solution. For the I^{133} determination, it was first necessary to find from the second extraction the amount of I^{132} interfering in the I^{133} region. Then, the quantity of I^{133} in the second extraction was calculated. The Bateman equation was used to derive the Te^{133m} content at time of first extraction. A straight extrapolation back to mid sampling time was made to obtain the Te^{133m} content of the air at that time. Application of the Bateman equation to the data from the first extraction gave a relationship between the I^{133} that existed in the first extraction, the initial I^{133} in the air at mid-sampling time, and the Te^{133m} in the air at mid-sampling time. With two quantities known, the I^{133} in the air at mid-sampling time was determined.

If most of the I^{131} had formed at the time of sampling, little or no I^{131} would appear in the second extraction. However, a significant amount was present, and its existence can be explained upon closer examination of the decay scheme (See Appendix, Part A). Early references list the ancestors of I^{131} as a 3.4-minute Sn and a 23-minute Sb. Current evidence points toward a 1.6-hour Sn as the fission fragment. Both values are reported as reliable, but neither one is listed as a metastable form. Also, Sb, the daughter of Sn, branches on decay to a 30-hour Te and a 24.8-minute Te. The 30-hour Te in turn branches on decay to the 24.8-minute Te and to a 8.08-day iodine.

The more significant parent of the I^{131} is the 24.8-minute Te^{131} which is formed from 85% of the Sb^{131} decay. Calculations indicate that at the H+2.5 mid-sampling time, only a small percentage (approximately 13%) of this Te^{131} had formed. Since it is the preponderant parent and is of shorter half-life, a small percentage of Te^{131} would imply an even smaller percentage of its daughter product. We therefore conclude that an insignificant amount of I^{131} was in the air at the time it was sampled. Absence of I^{131} in the physical samples supports this conslusion.

C-2. DETERMINATION OF IODINE IN DOG THYROIDS

Quantitative calculation of iodine in dog thyroids was based on the 0.53 Mev peak of I^{133} and the 0.36 Mev peak of the I^{131}. There is an interference from the 0.53 Mev peak in the 0.36 Mev region, but the contribution of the 0.36 Mev peak in the 0.53 Mev region is negligible. On this basis, and using the data from D+5, which indicated only two photopeaks, the I^{133} photopeak could be quantitated.

Data from D+22 provided information on the I^{131} for twelve of the thyroids. These quantities were extrapolated back to D+5 to compare them with the quantities calculated from the D+5 data. A ratio of the difference between the I^{131} values to the sum in the 0.53 Mev photopeak was determined to obtain the interference coefficient of the 0.53 Mev peak in the 0.36 Mev region. This ratio was determined for the twelve thyroids and an average value calculated. The average value for the interference coefficient was then used to determine the quantity of I^{131} in the remaining thyroids.

The amount of I^{131} and I^{133} determined in the thyroids was then extrapolated back to the time of sacrifice of each animal. This seems to be the only significant time at which the quantity of radioiodine measured by this method is meaningful. Before death, there is a build-up of iodine from decay of precursors in the other organs, and a decrease through biological elimination and through radioactive decay. After death radioactive decay is the only process continuing to affect iodine concentration in the thyroid.

The existence of I^{131} in some thyroids of the animals sacrificed in the field at the end of sampling time indicated that the dog thyroid is a more sensitive sampler for airborne iodine in a mixed fission product cloud than is standard air sampling equipment. This is due to the advantage provided by the living system's ability to concentrate and isolate iodine in the thyroid. This advantage seemed to hold even though the low volume equipment sampled from eight to forty times the amount of air the dogs inhaled. The I^{131} determinations were carried out twenty-two days after sacrifice to allow the shorter-lived iodines to decay out and permit observation of the I^{131}. The amounts remaining at D+22 were at the minimum detection limit, which probably explains why I^{131} was not observed in all thyroids counted.

The relatively long half-life of the Te^{132} (77.7 hours) would assure the virtual non-existence of its daughter I^{132} at the mid-sampling time of H+2.5. The rate of decay of I^{134} is such that by the time these samples

were assayed the isotope was not detectable. Quantitation of I^{135} was not attempted because of the multitude of photopeaks exhibited in the spectrum. Interference among the many photopeaks made any evaluation difficult and perhaps unrealistic.

PART D. SAMPLE CALCULATION OF PERCENTAGE Te^{131} IN AIR AT MID-SAMPLING TIME

The Bateman solution for a three-member chain is:

$$N_3 = \left[\frac{e^{-\lambda_1 t}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1 - \lambda_3)(\lambda_2 - \lambda_3)} \right] \lambda_1 \lambda_2 N_1^\circ$$

In this instance:

N_3 = Amount of Te^{131} at any time t.

N_1° = Initial quantity of Sn^{131}.

t = 2.5 hours, lapsed time after release.

$\lambda_1, \lambda_2, \lambda_3$ = Decay constants for Sn^{131}, Sb^{131}, and Te^{131}, respectively.

Numerical values for the decay constants are:

$\lambda_1 = 0.4431$

$\lambda_2 = 1.8078$

$\lambda_3 = 1.6632$

Numerical values for factors derived from the decay constants are:

$(\lambda_2 - \lambda_1) = 1.3647$

$(\lambda_3 - \lambda_1) = 1.2201$

$(\lambda_3 - \lambda_2) = 0.1446$

$e^{-\lambda_1 t} = 0.3430$

$e^{-\lambda_2 t} = 0.0116$

$e^{-\lambda_3 t} = 0.0164$

The solution then becomes:

$$N_3 = \left[\frac{0.3430}{(1.3647)(1.2201)} + \frac{0.0116}{(-1.3647)(-0.1446)} + \frac{0.0164}{(-1.2201)(0.1446)} \right] (0.4431)(1.8078) N_1^\circ$$

$$= 0.1306\, N_1^\circ = 13\%\, N_1^\circ$$